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Assessment of Heavy Metals, Gross Alpha, Gross Beta and Radon Activity Concentration in Groundwater around Doguwa and its Environs, within Kano State, Nigeria

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Abstract

This study assessed the activity concentrations of gross alpha, gross beta, Radon and heavy metals in groundwater sources from illegal mining areas in Doguwa Local Government, Kano State, Nigeria. Water samples from five distinct mining areas were analyzed for gross alpha and beta radioactivity using a portable single-channel gas-free proportional detector (MPC2000B-DP) via ISO9696 and ISO9697 methods, Radon levels with a Rad7 (DURRIDGE) detector, and heavy metals with atomic absorption spectroscopy. Gross alpha concentrations ranged from 0.0000238 to 0.00013 Bq/L, averaging 0.00008158 Bq/L, a value below the WHO limit of 0.5 Bq/L. Gross beta concentrations ranged from 0.536 to 2.78 Bq/L, averaging 1.7056 Bq/L, exceeding the WHO limit of 1.0 Bq/L. Radon levels varied from 0.12 to 1.7 Bq/L, averaging 1.102 Bq/L, below the WHO limit of 11.1 Bq/L. Annual effective doses from gross alpha ingestion were 4.10918E-05, 4.16874E-06, and 8.33748E-06 mSv/year for adults, infants, and children, respectively. Radon exposure doses were 1.61E-13 and 8.04E-14 mSv/year for adults and children, respectively, below the 0.1 mSv/year limit. However, beta radiation doses exceeded the 0.1 mSv/year limit, with values of 0.85911072, 0.21477768, and 0.42955536 mSv/year for adults, infants, and children, respectively. The average concentrations of heavy metals were 0.00058 mg/L for Cd, 0.012 mg/L for Cr, 0.00628 mg/L for Fe, 0.0046 mg/L for Mn, 0.09534 mg/L for Ni, 0.01214 mg/L for Pb, and 0.00582 mg/L for Zn. Children exhibited elevated cancer risks from heavy metal ingestion 0.004865 and dermal exposure 0.000069, with hazard quotient values of 0.740205 and 0.004882, respectively. The hazard index and lifetime cancer risk for children were 0.004882, exceeding USEPA recommended values. While gross alpha and radon levels were within safety limits, beta radiation levels and heavy metal hazard quotients exceeded maximum contamination levels, highlighting significant health risks, including DNA damage and increased cancer risks.

Keywords: Annual effective dose; Excess life cancer risk; Gross alpha; Gross beta; Radon.

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I. INTRODUCTION

Water quality is an important parameter of environmental studies. Radioactivity in surface water is mainly due to radioactive elements in the earth's crust. The earth's crust contains small amounts of Uranium, Thorium and Radium, and radioactive isotope of Potassium[1].

Gross alpha and beta radioactivity in water may increase the long-term incidence of cancer and many other health hazards, especially when these radionuclides are deposited in the human body through ingestion or inhalation. Surface and groundwater resources in mining vicinities may be contaminated with alpha emitters such as ^{238}U, ^{234}U, ^{232}Th, ^{226}Ra and ^{210}Po and beta emitters such as ^{40}K, ^{228}Ra and ^{210}Pb due to the large volume of radioactivity bearing excavated tailings. [2].

Radon is a colourless and odourless radioactive gas that exists naturally in soil, air, and groundwater. It exists in three different isotopes in the ^{238}U, ^{232}Th and ^{235}U decay series respectively. Due to the relative forms as ^{222}Rn ($t_{1/2} = 3:8\ d$), ^{220}Rn ($t_{1/2} = 55:6\ s$) and ^{219}Rn ($t_{1/2} = 4\ s$), short half-lives of the latter two radon isotopes, ^{222}Rn is mostly given significant consideration from a radiological health hazard perspective [3].

The concentration of radionuclides, heavy metals, and other poisonous microbes or infectious agents in water can assume hazardous proportions under certain environmental matrixes, which can result in both carcinogenic and non-carcinogenic risks [4]. The major exposure pathways are through inhalation, ingestion, and thermal contact or absorption [5].

Research has been conducted on the investigation of the activity concentrations of heavy metals, gross alpha, gross beta, and radon in groundwater, and the evaluation of the corresponding annual effective cancer risk in humans, both within and outside Nigeria. Recently, [6] conducted a study on the assessment of Radon concentration in groundwater with associated human health implications around Bagwai and Shanono artisan gold mining sites in Kano State, North-western Nigeria. Reference [7], also carried out a study on the determination of the concentration of heavy metals and Radon in soil and water samples from Wadi-B Jere oil exploration sites in Maiduguri, Northeast Nigeria, while the presence of heavy metals in water samples within the southern part of Kaduna State, Nigeria were determined in a different study carried out by [8] using the Atomic Absorption Spectrophotometer (AAS). Similarly, in a research conducted by [9], gross alpha and beta activity concentrations were determined alongside the estimated adult and infant dose intake in surface and groundwater of ten (10) oil field environments in the western Niger Delta of Nigeria using gas-flow proportional counters. Also, [10] estimated the Radon concentration in groundwater and soil samples from the Riruwai Mining site, using a DURRIDGE RAD7 electronic radon detector. Another research conducted by [3] determined radon mapping and assessment of health risks from heavy metals in drinking water in southwest Nigeria, while [11] evaluated the concentration of heavy metals and natural gross radioactivity measured in the surface water and sediment of Hazar Lake, Elazıg, Turkey. Reference [12], also determined the concentration of heavy metals in water and sediment in the river Enumabia in Orokam community, Benue State, Nigeria, using an AA-700 Dual Atomizer Atomic Absorption Spectrophotometer. Correspondingly, [13] determined gross alpha and beta activity concentrations of eighteen water samples, consisting of boreholes and Hand-dug wells from Sabon Gari Local government area of Kaduna State, Nigeria, using the potable single channel gas-free MPC-2000B-DP detector.

There is currently no data on the levels of gross alpha, gross beta, Radon and heavy metals in the groundwater of Doguwa and its surroundings. However, a reconnaissance survey revealed that illegal mining of tin, gold, and uranium has occurred within the research areas for a decade, using rudimentary tools and unregulated methods. This activity probably makes the surrounding water radioactive, posing health risks, thus highlighting the urgent need for a study to measure these concentrations.

II. MATERIALS AND METHODS

A. Location of the Study Area

Doguwa Local Government Area (LGA) in Kano State, Nigeria shares borders with several other LGAs, Tudun Wada located to the west, Dawakin Kudu situated to the northwest, Ungogo located to the north, Gezawa situated to the northeast, Minjibir located to the east, and Rogo situated to the southeast. The area under study lies within latitude 11.75^{0}N and longitude 8.52^{0}E and houses the largest underground tin mining sites in Kano, north-western Nigeria, which were officially closed in 1984 and have been reopened by artisanal and illegal miners thereafter. The areas have been identified as one of the major potential uranium mining sites in Nigeria. Many minerals, such as lead, and thorium, are reported to be on a commercial scale, besides tin, which is mined daily [14].

B. Materials

The materials used for this study include 2-liter plastic containers, Beakers (Pyrex), Gloves, Blunt forceps,
an Oven, a Hot plate, an Analytical balance, a Spatula, a Fume cupboard, a Petri dish (crucible), a Planchet, a Police-man (rubber), an MPC 2000 B-DP (Dual Phosphor), syringes and needles, GPS device, Gas free proportional Counter, Rad7 Detector, atomic absorption spectrometer (AAS).

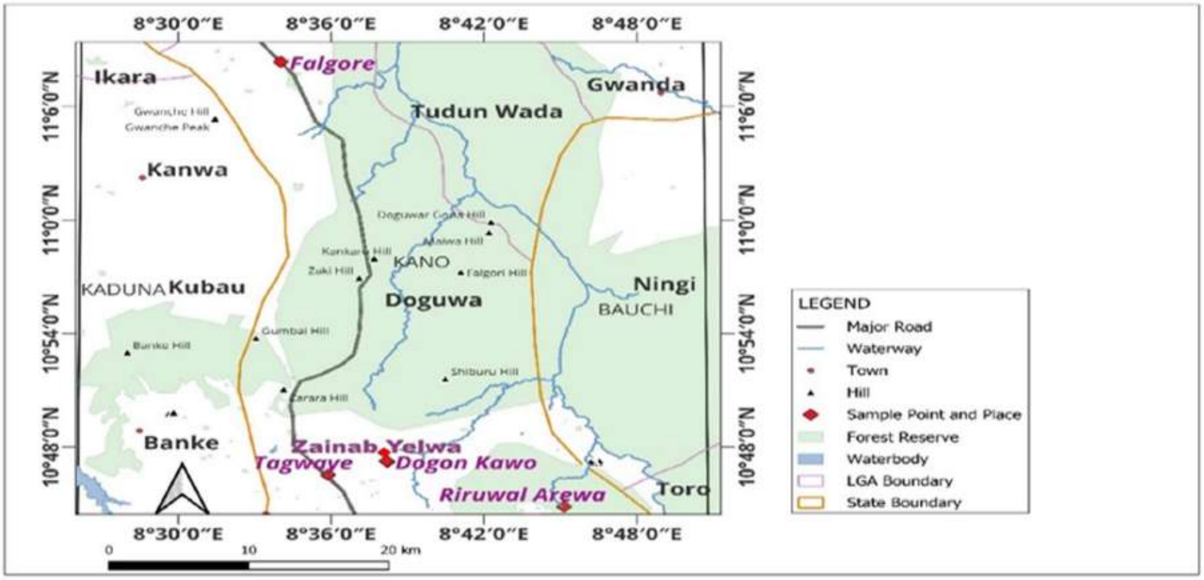


Fig. 1 Geographical map of the study area.

Fig. 2 RAD7 detector.

Fig. 3 MPC2000B-DP (machine for analysis gross alpha & beta).

C. Methods

The international standards organization procedure (ISO9696 and ISO 9697: 1992E) for the measurement of gross alpha and gross beta activity in water was employed in this analysis. This method provided a screening technique to measure the gross alpha and beta radioactivity in water samples while for Radon and heavy metals, a systematic approach was employed for the collection of groundwater samples specifically from five distinct mining sites areas.

1) *Sample Preparation for Gross Alpha and Gross Beta*

The water samples collected were preserved under the ISO standard (20 mL of 50% V/V of HNO_3 per litre of water). The purpose of this is to minimize the loss of radioactive material from the solution due to absorption. Out of the two litres of each water sample collected, some of it was evaporated to a decreased volume of 100-50 mL using a hot plate. It was transferred into a weighed porcelain dish or petri dish of 150 mL and placed under an infra-red lump until it was dried completely, and it was left to cool inside the desiccator to normal condition and weighed as m_2. The weight of the porcelain dish was subtracted from m_2 to obtain the weight of the residue (m_r) in milligrams. If the weight of the residue is greater than the required residue, then only the required residue size is taken into a weighed counting planchet. The reagent (Vinyl acetate) is added, and the source is distributed evenly on the surface of the planchet. Each sample was counted for 45 min (2700 s) for 5 cycles and the average result was taken.

2) *Sample Preparation for Radon*

A systematic approach was employed for the collection of groundwater samples specifically from wells in the mining sites areas. Each sampling point near the mining areas involved the collection of 2 litres of water in meticulously cleaned bottles, the samples were sealed with the provided air-tight sample vial caps and thereafter transported to the laboratory for analysis.

The samples were analyzed using the Durridge Inc. Rad 7 detector which is a sophisticated and active radon detector. This device uses a solid-state detector (usually Silicon) that converts alpha radiation directly into an electrical signal. The system determines ^{222}Rn and ^{220}Rn (thoron) activity concentration released into continuously circulating air (in a closed loop), which is in equilibrium with a constant stream of water passing through an air-water exchanger. To analyse radon in water, an accessory RADH2O is connected to the Rad 7, which uses a computer-driven electronic detector with pre-programmed setups that print out a summary showing the average radon reading in 30 minutes. The RAD7/RADH2O system is well documented [15].

3) Sample analysis for heavy metals

The samples were digested to liberate metals into a solution and fed into an atomic absorption spectrometer, where the metals are vaporized and subjected to light at precise wavelengths. The degree of light absorption by the metals correlates directly with their concentration in the sample. By employing a calibration curve, the metal concentrations are precisely quantified. Rigorous quality control protocols are applied to guarantee result accuracy [7].

D. Health Risk Assessment for Gross Alpha and Beta

1) Estimation of Annual Effective Dose due to alpha and beta

The Annual Effective Dose Equivalent (AEDE) is the quantity of ionizing radiation a person may receive in a year according to protection guidelines. The formula for computation of AEDE for gross alpha or gross beta radiation received is given in (1).

$$AEDE_{\alpha,\beta} = A_{\alpha,\beta} \times IR_w \times DCF_{\alpha,\beta} \quad (1)$$

Where, $AEDE_{\alpha,\beta}$ = Annual Effective Dose Equivalent, $DCF_{\alpha,\beta}$ = Dose Conversion Factor (mSv/Bq), $A_{\alpha,\beta}$ = Activity (Bq/L), IR_w = Intake of water for an adult in a year (2 L/day) = 730 Liters, for an infant (≤1 yr) in a year is 182.5 L, and for a child (1–12 yrs) in a year is 365 L, Dose Conversion Factors (DCF) for ^{226}Ra (gross alpha) is 2.80×10^{-4}mSv Bq^{-1} and for ^{228}Ra(gross beta) is 6.90×10^{-4} mSvBq^{-1} as published by the WHO [2], [14].

2) Excess Lifetime Cancer Risk (ELCR) due to Gross Alpha and Beta

Excess Lifetime Cancer Risk (ELCR) is the probability of developing cancer over a lifetime at a given exposure level. In this work, 70 years was considered as the average duration of life for humans. ELCR for gross alpha or gross beta was calculated using (2).

$$ELCR_{\alpha,\beta} = AEDE \times DL \times RF \quad (2)$$

Where DL is the average life span of a man (estimated to be 70 years), and RF is the Risk Factor (Sv^{-1}), which is also the fatal cancer risk per Sievert. For stochastic effects, the International Commission on Radiological Protection (ICRP) recommended RF as 0.05 Sv^{-1} equivalent to 5.0×10^{-5} (mSv^{-1}) for the public [16].

E. Health Risk Assessment for Radon

1) Determination of Radon concentration

The ^{222}Rn concentration in a sample of water can be determined using (3) as proposed by [7].

$$Rn\ (Bql^{-1}) = \frac{100-(SC-BC)exp\ (\lambda t)}{60(CF)} \quad (3)$$

Where, Rn = Radon concentration in Bql^{-1}, SC = Sample count rate (count/min), BC = Background count rate (count/min), t = Elapsed time from sampling to testing given in minutes, CF = Calibration factor and D = Decay time.

2) Estimation of Annual Effective Dose Due to Radon

The AED (mSv) was calculated using (4).

$$AED\ (mSv) = K \times G \times C \times t \quad (4)$$

Where, K is the ingesting dose conversion factor of ^{222}Rn (10^{-8} SvBq^{-1} for adults and 2×10^{-8} SvBq^{-1} children respectively), G, is the water consumption per day (2 litres/day and 1 litre/day for adults and children respectively), C, is the concentration of ^{222}Rn (Bq/L), t, the period of consumption, (365 days or 1 year) [15].

3) Excess Life Cancer Risk due to ingestion of Radon

Radiation dose due to ingestion for different age categories was calculated using (5) to determine the annual effective dose.

$$ELCR = AEDE \times DL \times RF \quad (5)$$

Where AEDE is the annual effective dose mSv/y, DL is the life expectancy (70 years), and RF is the fatal risk factor per Sievert (Sv). In the case of stochastic effects, ICRP-60 uses an RF of 0.05 for the public[7].

F. Health Risk assessment for Heavy metals

1) Chronic daily intake (CDI)

Ingestion and dermal absorption, the most common and important exposure pathways for water in the living environment is selected for human health risk assessment. The US Environmental Protection Agency (USEPA) points out that the number of pollutants absorbed by the human body is calculated based on chronic daily intake (CDI) given by (6).

$$\mathrm{CDI_{ing}} = \frac{\mathrm{C \times IR \times EF \times ED}}{\mathrm{AT \times BW}} \quad (6)$$

$\mathrm{CDI_{ing}}$ is the chronic daily intake of heavy metals ingested in mg/kg-day, IR = 1.277 Ld^{-1} is the ingestion rate, C = concentration of heavy metals in mg/kg, EF = 365 days is the exposure frequency, ED = 74.8 a, is exposure duration in years, BW = 63.1 kg is the body weight of the exposed individual, AT = 27302 d is period, and CF = 10^{-6} Lcm^{-3} is the conversion factor [17].

For Chronic daily intake for dermal absorption is given by (7).

$$CDI_{derm} = \frac{C\times SA\times KC\times ABS\times ET\times EF\times ED}{AT\times BW} \quad (7)$$

Where C is the heavy metal concentration (mg/L) in water; EF is exposure frequency: 365 d/y (USEPA, 2020); IR is ingestion rate: 2 L/d for adult and 1 L/d for children; ED is exposure duration: 70 years for adult and 6 years for the children, BW is body weight: 60 kg for adult, 15 kg for children, AT is average time: 25,550 days for adult and 2190 days for children, Skin surface area (SA) for water exposure are 5700 cm^2/d (adult), 2800 cm^2/d (child), KC is dermal permeability factor: 0.001 for As and Cd, 0.002 for Cr and 0.004 for Pb cm/h, ET is exposure time: 0.8 h/d for adult and 0.6 h/d for children; 10 6 was used to convert from kilo- gram to milligram; ABS is fraction of dermal absorption: 0.03 (for As) and 0.001 (for others) [18].

2) Hazard quotient (HQ)

For the assessment of non-carcinogenic health hazards, the hazard quotient (HQ) was utilized. The parameter evaluates the risk from ingestion and dermal absorption of each heavy metal in water and is expressed in (8).

$$HQ = \frac{CDI}{RfD} \quad (8)$$

Where *RfD* is the reference dose for heavy metal (mg kg $-^1$ day $-^1$).

The non-carcinogenic effect is considered insignificant if HQ < 1. A value greater than 1

Where CDI is the chronic daily intake (mg $kg^{-1} d^{-1}$); RfD is the chronic reference dose values of heavy metals (mg per kg per day): 0.0005 (RfD_{ing}) and 0.000025 (RfD_{derm}) for Cd; 0.0003 (both RfD_{ing} and RfD_{derm}) for As; 0.003 (RfD_{ing}) and 0.000075 (RfD_{derm}) for Cr; 0.0014 (RfD_{ing}) and 0.00042 (RfD_{derm}) for Pb [17].

3) Hazard Index

Using the Hazard Index, one can assess the overall non-carcinogenic health risk exposure of dictated heavy metals emanating from different pathways. For this study, HI is expressed as the summation of the individual hazard quotient of the measured heavy metals for both ingestion and dermal absorption routes as given in (9) [18].

$$HI = \sum HQ_{ing} + HQ_{derm} \quad (9)$$

HI < 1 implies an insignificant non-carcinogenic risk to the exposed public.

4) Cancer Risk (CR)

The cancer risk (CR) due to the ingestion of heavy metals in water to human health is estimated by the multiplication of the CDI ($mg.kg^{-1}.day^{-1}$) and the cancer slope factor (CSF) (mg^{-1}.kg.day), as in (10).

$$CR = CDI \times CSF \quad (10)$$

Where CR is the cancer risk over a lifetime by heavy metals. The acceptable levels of CR values should fall within 10^{-6} and 10^{-4}.

5) Lifetime Cancer Risk (LCR)

The lifetime cancer risk (LCR) was calculated from the cancer risk from both ingestion and dermal absorption pathways. The LCR was computed using (11).

$$LCR = \sum CR_{ing} + CR_{derm} \quad (11)$$

Where CR_{ing} and CR_{derm} represent the carcinogenic risks from ingestion and dermal absorption pathways [18].

III. RESULTS AND DISCUSSION

A. Result and Discussion for Gross Alpha

Table I presents the concentration of gross alpha (Bq/L), annual effective dose (mSv/year), and excess lifetime cancer risk (mSv/year) in adults, infants, and children which are evaluated using (1) and (2) respectively. The overall arithmetic mean gross alpha activity concentration in the groundwater water evaluated was 0.00008158 Bq/L, with the highest value recorded at R/Arewa and the lowest at D/Kawu. All the values are less than the maximum permissible limit of 0.5Bq/L set by WHO. Fig. 4 shows the comparison with the recommended value set by the World Health Organization (WHO).

The arithmetic mean values for excess lifetime cancer risk due to gross alpha radiation for adults, infants, and children are 0.000000143821, 0.0000000145906, and 0.000000029181 mSv/year, respectively. All values are well below the maximum permissible limit of 0.29 mSv/year. This indicates that the likelihood of contracting cancer from the reported contamination is low, given the low activity concentration of gross alpha. The activity concentration of gross alpha, annual effective dose, and excess lifetime cancer risk in different age groups are illustrated in Fig. 5.

Table I. Gross alpha Concentration, Annual Effective Dose and Excess Life Cancer Risk.

Sample I.D	Gross Alpha (Bq/L)	AED(α) Adult	AED(α) Infant	AED(α) Child	ELCR (Adult)	ELCR (Infant)	ELCR (Child)
R/Arewa	0.00013	6.5481E-05	6.6430E-06	1.32860E-5	2.29184E-07	2.32505E-08	4.6501E-08
Z/Yelwa	0.000127	6.39699E-05	6.489.7E-06	1.29794E-05	2.23895E-07	2.2714E-08	4.5428E-08
D/Kawo	0.0000477	2.40265E-05	2.43747E-06	4.87494E-06	8.40927E-08	8.53115E-09	1.7062E-08
Tagwaye	0.0000794	3.99938E-05	4.05734E-06	8.11468E-06	1.39978E-07	1.42007E-08	2.8401E-08
Falgore	0.0000238	1.19881E-05	1.21618E-06	2.43236E-06	4.19582E-08	4.25663E-09	8.5133E-09
Average	0.00008158	4.10918E-05	4.16874E-06	8.33748E-06	1.43821E-07	1.45906E-08	2.9181E-08

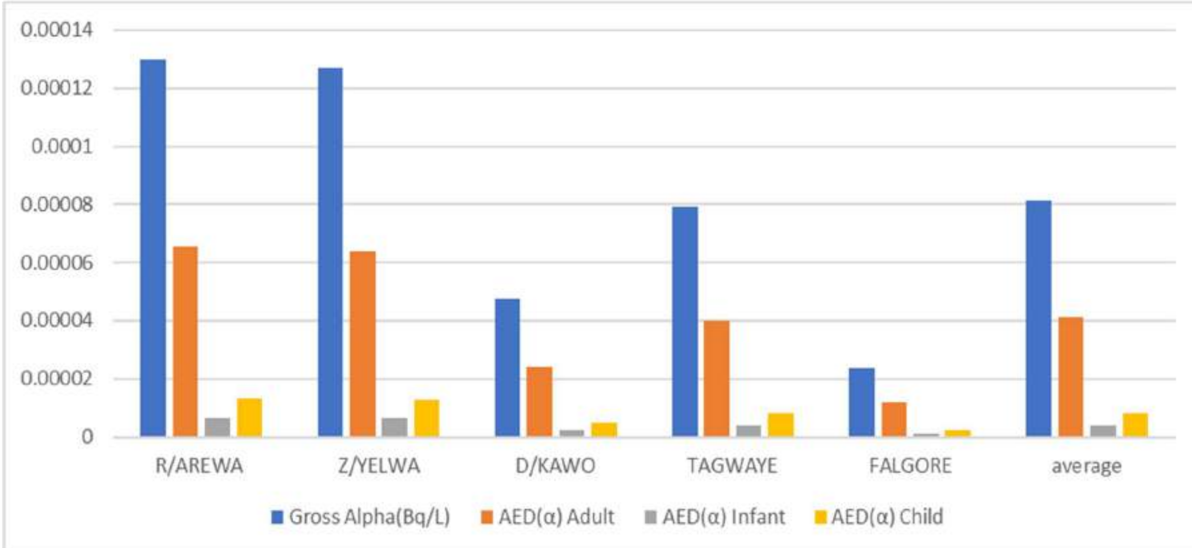
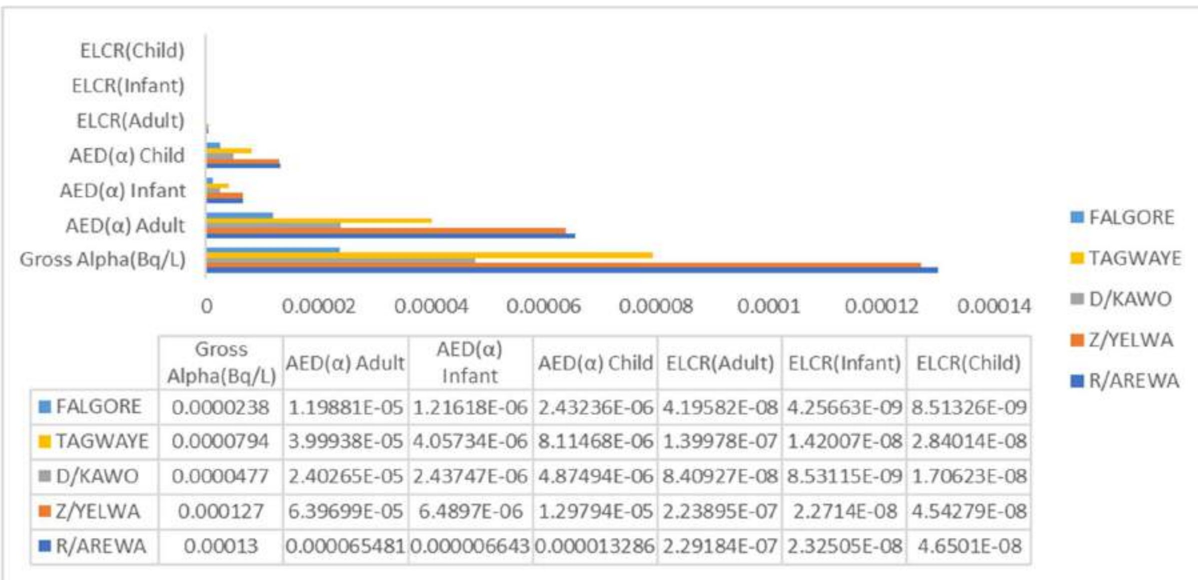


Fig. 4 Gross alpha, AED and ELCR concentration on adult, infant and child in the Study Area.



	Gross Alpha(Bq/L)	AED(α) Adult	AED(α) Infant	AED(α) Child	ELCR(Adult)	ELCR(Infant)	ELCR(Child)
FALGORE	0.0000238	1.19881E-05	1.21618E-06	2.43236E-06	4.19582E-08	4.25663E-09	8.51326E-09
TAGWAYE	0.0000794	3.99938E-05	4.05734E-06	8.11468E-06	1.39978E-07	1.42007E-08	2.84014E-08
D/KAWO	0.0000477	2.40265E-05	2.43747E-06	4.87494E-06	8.40927E-08	8.53115E-09	1.70623E-08
Z/YELWA	0.000127	6.39699E-05	6.4897E-06	1.29794E-05	2.23895E-07	2.2714E-08	4.54279E-08
R/AREWA	0.00013	0.000065481	0.000006643	0.000013286	2.29184E-07	2.32505E-08	4.6501E-08

Fig. 5 Shows the average values for Gross alpha, AED for gross alpha, and ELCR for gross alpha on adults, infants and children in the study area.

B. Gross Beta Result analysis and discussion

Table II outlines the concentration of gross beta (Bq/L), annual effective dose (mSv/year), and excess lifetime cancer risk (mSv/year) for adults, infants, and children which are evaluated using (1) and (2) respectively. The arithmetic mean gross beta activity concentration in the drinking water was calculated at 1.7056 Bq/L. Notably, this value exceeds the maximum permissible limit set by the World Health Organization (WHO), which stands at 1.0 Bq/L. The recorded range of beta concentration spans from its highest in R/Arewa to its lowest in Tagwaye, as depicted in Fig. 6.

The data depicted in Fig. 7 underscore a concerning trend. All values for gross beta, annual committed effective dose, and excess lifetime cancer risk for the age groups surpass the permissible dose contribution from water. This is particularly worrisome as it indicates a significant impact of beta radiation on adults, infants, and children. Beta radiation, known for its ability to penetrate tissues and potentially cause internal harm upon ingestion, poses elevated risks of developing various cancers, including leukaemia and bone cancer.

Table II: Gross Beta Concentration, Annual Effective Dose and Excess Life Cancer Risk.

Sample ID	Gross Beta (Bq/L)	AED(β) Adult	AED(β) Infant	AED(β) Child	ELCR (Adult)	ELCR (Infant)	ELCR (Child)
R/Arewa	2.78	1.400286	0.3500715	0.700143	0.004901001	0.00122525	0.002450501
Z/Yelwa	2.53	1.274361	0.31859025	0.6371805	0.004460264	0.001115066	0.002230132
D/Kawo	0.682	0.3435234	0.08588085	0.1717617	0.001202332	0.000300583	0.000601166
Tagwaye	0.536	0.2699832	0.0674958	0.1349916	0.000944941	0.000236235	0.000472471
Falgore	2.00	1.0074	0.25185	0.5037	0.0035259	0.000881475	0.00176295
Average	1.7056	0.85911072	0.21477768	0.42955536	0.003006888	0.000751722	0.001503444

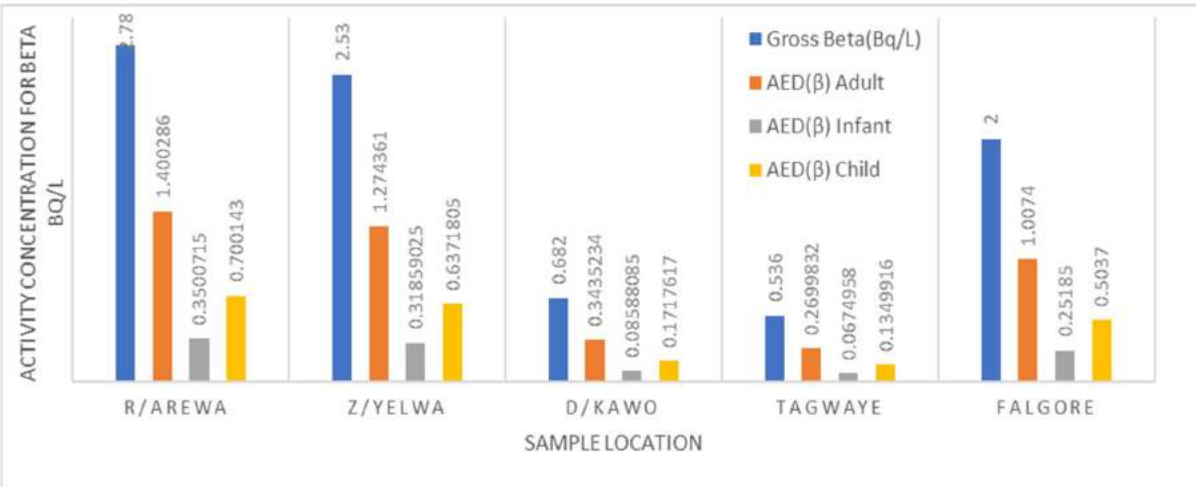


Fig. 6 Gross beta, AED and ELCR concentration on adults, infants and children in the study area.

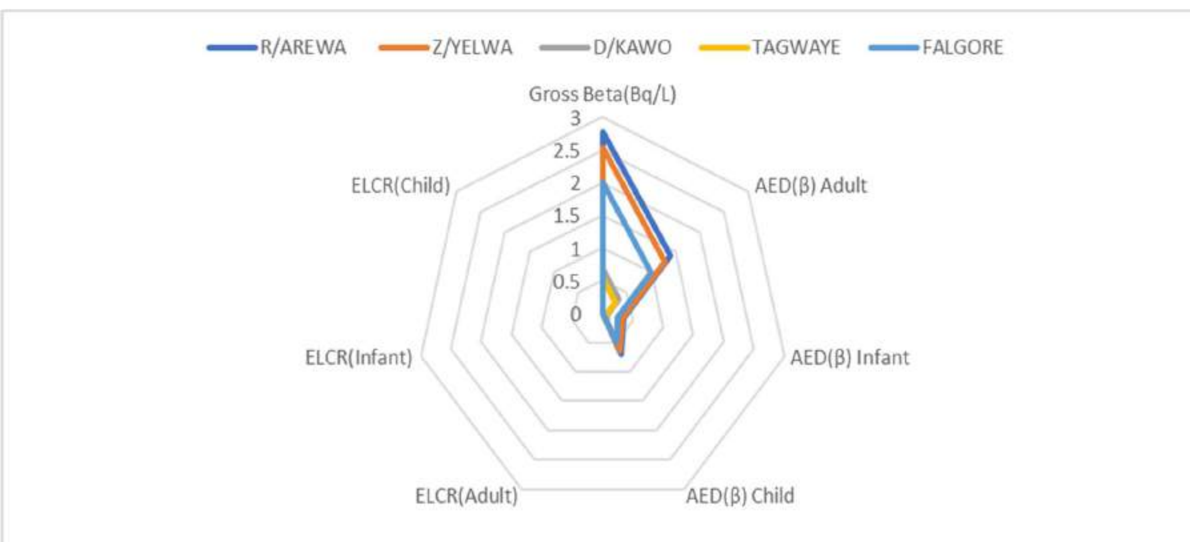


Fig. 7 Distribution of Gross beta, ELCR and AED for different age groups in the study area.

C. Comparison Between Gross Alpha and Beta

Fig. 8 compares the activity concentrations of gross alpha and beta radiation, revealing that the gross beta activity concentration is the highest across all the mining areas and exceeds the recommended limit of 1.0 Bq/L. Fig. 9 illustrates the average activity concentrations of gross alpha and beta, clearly indicating that the average beta concentration is significantly higher than that of alpha. This suggests that the water is contaminated by beta radionuclides, exceeding the average recommended value. Fig. 10 shows the comparison of alpha and beta with recommended values set by WHO, indicating that R/Arewa has the highest concentrations of both alpha and beta radiation, while Tagwaye has the lowest beta concentration and D/Kawu has the lowest alpha concentration.

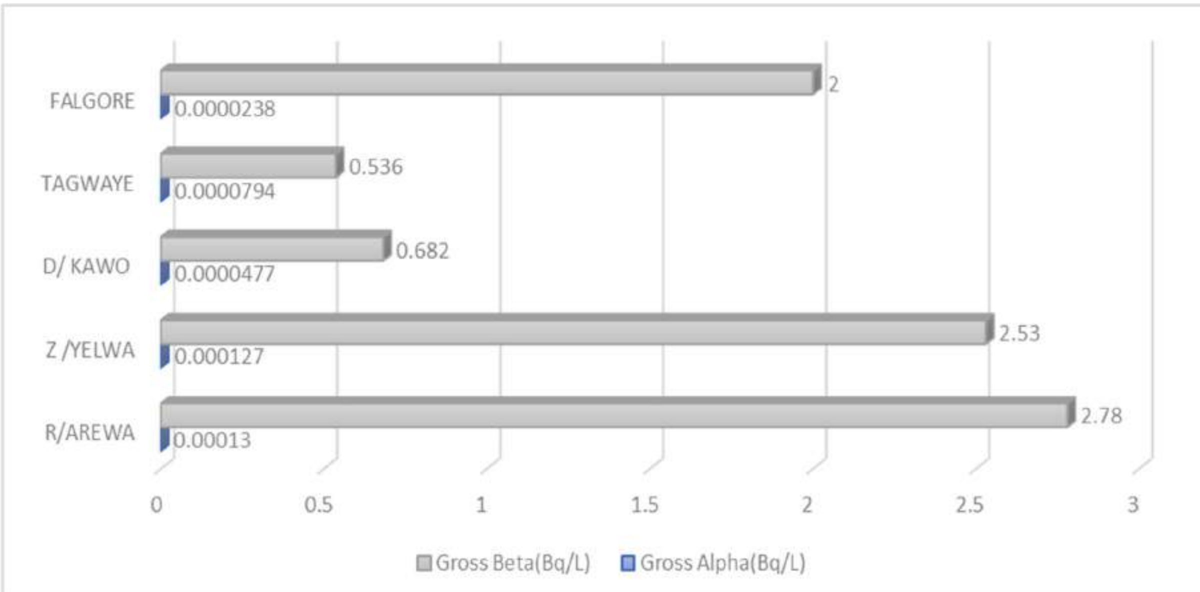


Fig. 8 Comparison of activity concentration of gross alpha and beta in the study areas.

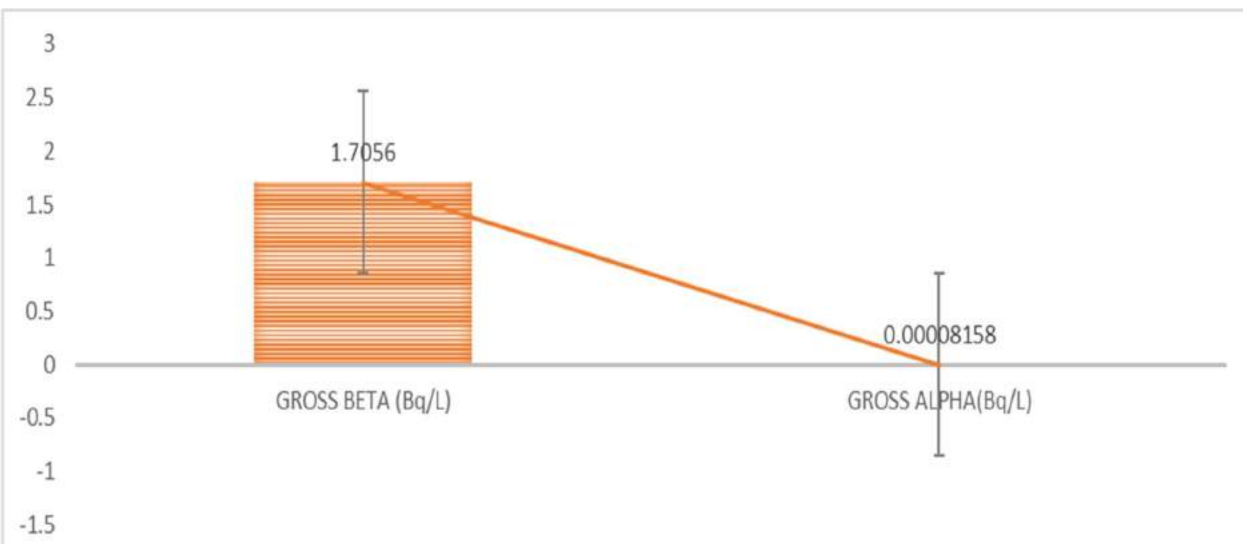


Fig. 9 Comparison of average activity concentration of gross alpha and beta in the study area.

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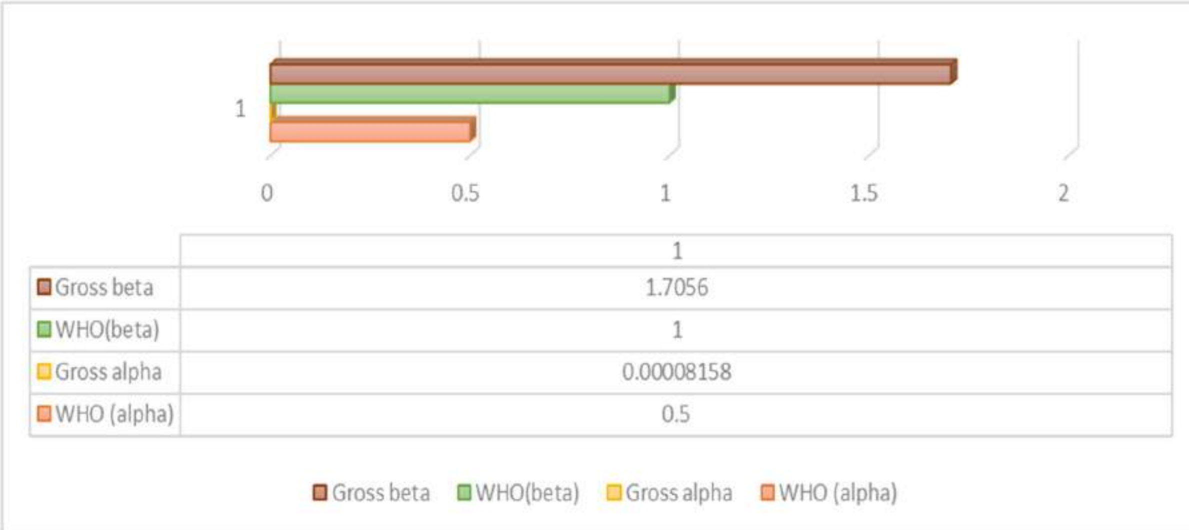

Fig. 10 Comparison of Gross alpha and Gross beta Concentration (BqL^{-1}) in the study area WHO recommended limit.

D. Radon Result Analysis and Discussion

Table III presents the results of Radon activity concentration, annual effective dose, and excess lifetime cancer risk due to ingestion of radon in water samples calculated using (3), (4), and (5). The Radon activity concentration in the water samples ranged from 0.12 to 1.7 Bq/L, with a mean value of 1.102 Bq/L. The highest radon activity concentration was recorded in Dogon Kawu, while the lowest was found in Falgore, as illustrated in Fig. 11. The mean Radon activity concentration of 1.102 Bq/L in water from the study area indicates that the Radon activity concentration is significantly lower than the Maximum Contaminant Level (MCL) set by the United States Environmental Protection Agency (USEPA), which is 11.1 Bq/L. Additionally, when compared to the standard value of 100 Bq/L recommended by the European Commission n (EC), the radon activity concentration in all samples remained well below the recommended limit.

The annual effective dose values for adults and children ranged from 1.75E-14 to 2.48E-13 mSv/y, with an average value of 1.61E-13 mSv/y for adults and from 8.76E-15 to 1.15E-13 mSv/y with an average value of 8.04E-14 mSv/y for children. The excess lifetime cancer risk (ELCR) in adults and children, due to effective doses resulting from 222Ra intake in water samples, ranged from 6.13E-14 to 8.69E-13 mSv/y with an average of 8.69E-13 mSv/y for adults, and from 3.07E-14 to 2.07E-13 mSv/y with an average value of 2.82E-13 mSv/y for children. These values are significantly below the International Commission on Radiological Protection (ICRP) recommended limit of 1 mSv/y for the ingestion of radionuclides in drinking water by the public for prolonged exposure. Fig. 12 illustrates the comparison of excess lifetime cancer risk between children and adults. Fig. 13 shows that the ELCR due to radon is 67% for adults and 33% for children.

Table III. Radon Concentration, Annual Effective Dose and Excess Life Cancer Risk

Location	Radon Conc. (Bq/L)	AED (adult)	AED (child)	ELCR (adult)	ELCR (child)
R/Arewa	1.57	2.29E-13	1.15E-13	8.02E-13	4.01E-13
Z /Yelwa	1.31	1.91E-13	9.56E-14	6.69E-13	3.35E-13
D/ Kawo	1.7	2.48E-13	1.24E-13	8.69E-13	4.34E-13
Tagwaye	0.81	1.18E-13	5.91E-14	4.14E-13	2.07E-13
Falgore	0.12	1.75E-14	8.76E-15	6.13E-14	3.07E-14
Average	1.102	1.61E-13	8.04E-14	5.63E-13	2.82E-13

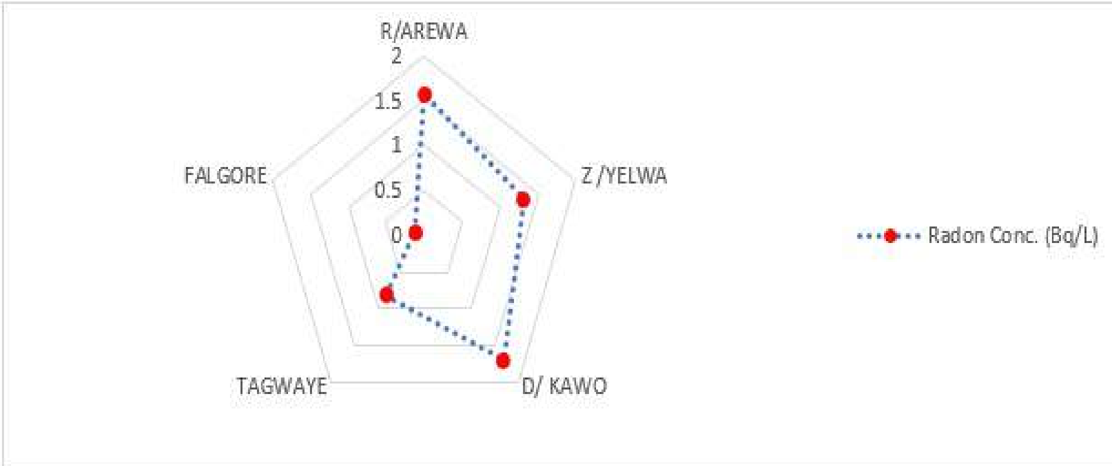

Fig. 11 Radon Concentration (BqL^{-1}).

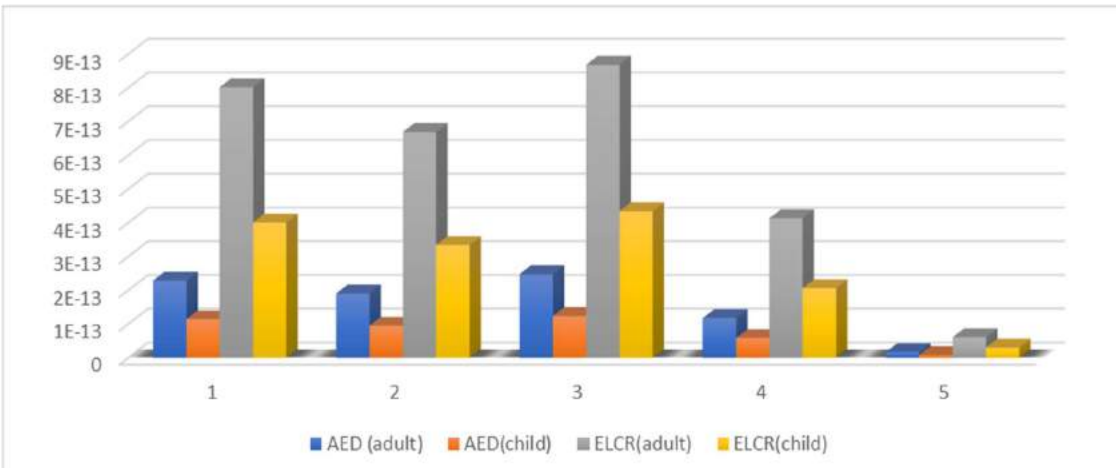

Fig. 12 AED and ELCR on adults and children in the study areas.

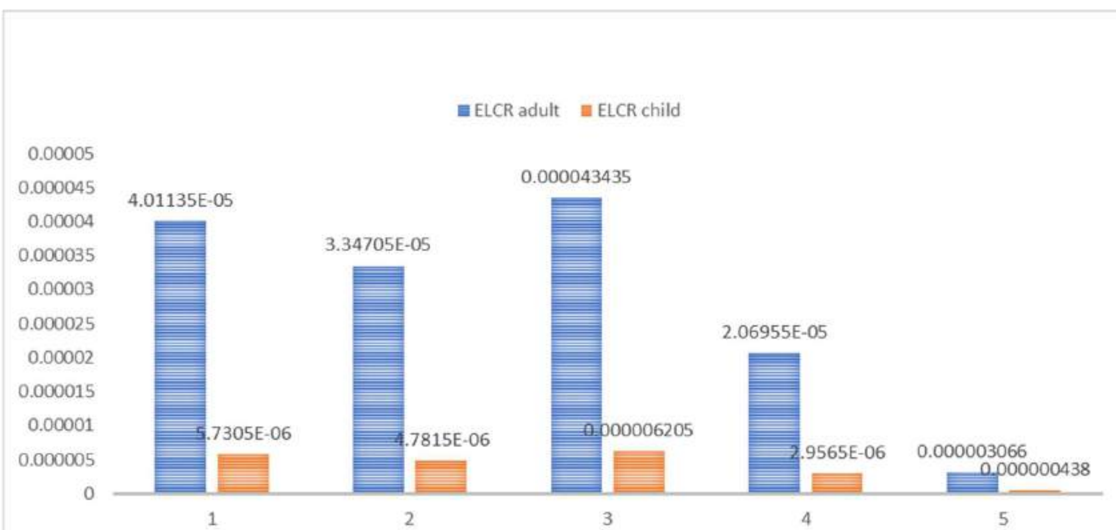

Fig. 13 Comparison of excess life cancer risk of child and adult.

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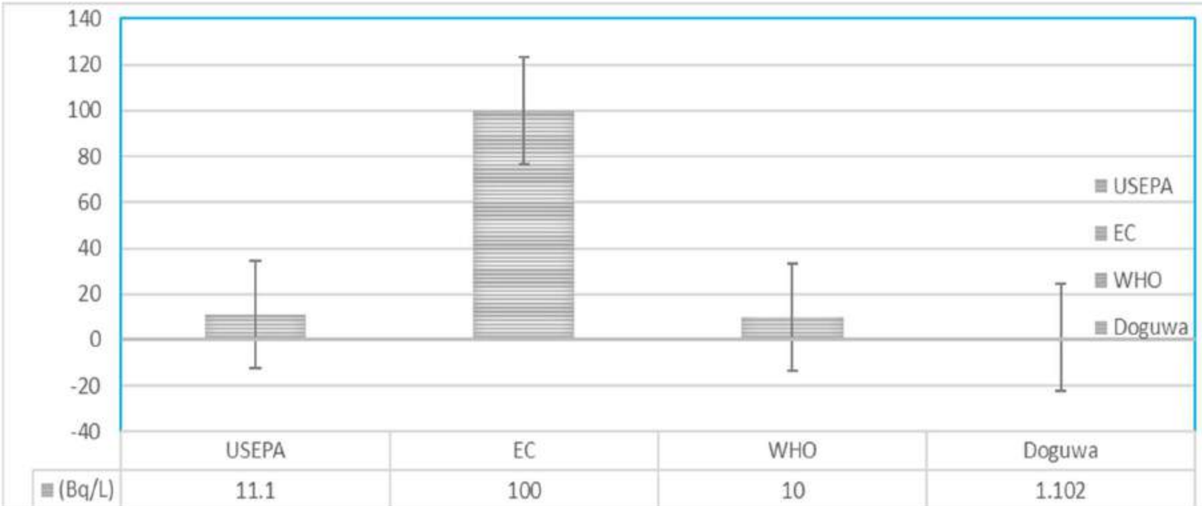

Fig. 14 Comparison of Radon Concentration (BqL^{-1}) in the Study Area with USEPA, EC and WHO Recommended Limit.

E. Heavy Metals Result and Discussion

Table IV presents the results of heavy metals activity concentration. The average heavy metals activity concentrations in the water samples are 0.00058 mg/L for Cd, 0.012 mg/L for Cr, 0.00628 mg/L for Fe, 0.0046 mg/L for Mn, 0.09534 mg/L for Ni, 0.01214 mg/L for Pb, and 0.00582 mg/L for Zn. The highest concentration of heavy metals was found to be Nickel in R/Arewa, while the lowest was Cadmium in D/Kawo. The activity concentrations are in the order of Cd < Mn < Zn < Fe < Cr < Pb < Ni.

It was found that the average values of all the heavy metals are below the recommended values set by the WHO. Fig. 15 shows a comparison of the recommended values set by WHO with the average concentrations of each heavy metal.

The average chronic daily intake (CDI) for adults, both through ingestion and dermal exposure, was determined to be 0.42514 and 2.88E-05, respectively, using (6) and (7). The hazard quotient (HQ) for both ingestion and dermal exposure was found to be 2.88E-05, calculated using (8). The hazard index (HI) for adults was 0.426802, as determined by (9). Additionally, the lifetime cancer risk (LCR) for adults was calculated to be 0.3333 using (10).

The average HQ for both ingestion and dermal exposure in adults is significantly higher than the recommended value. The LCR values also indicate a high potential risk, suggesting that there is a substantial health risk to adults using the water in the area. Fig. 16 shows the comparison of the hazard quotient and life cancer risk in comparison with the recommended value set by USEPA.

The average chronic daily intake (CDI) for children, through both ingestion and dermal exposure, was calculated to be 0.006247 and 1.85E-05, respectively, using (6) and (7) (see Table VI). The hazard quotient (HQ) for ingestion and dermal exposure was found to be 12.49361 and 0.740205, respectively, calculated using (8). The hazard index (HI) for children was determined to be 0.006266, as per (9). Additionally, the lifetime cancer risk (LCR) for adults was calculated to be 0.004882 using (10).

Table IV. Concentration of Heavy metals, Average and Recommended WHO values.

Location	Cd (mg/l)	Cr(mg/l)	Fe(mg/l)	Mn(mg/l)	Ni(mg/l)	Pb(mg/l)	Zn(mg/l)
R/Arewa	0.0009	0.0173	0.0051	0.006	0.3333	0.0075	0.0197
Z/Yelwa	0.0003	0.0089	0.0102	0.0013	0.0053	0.0119	0.0011
D/Kawo	0.0008	0.0107	0.0051	0.0076	0.0754	0.0159	0.003
Tagwaye	0.0003	0.0088	0.0061	0.0029	0.0491	0.0208	0.0014
Falgore	0.0006	0.0143	0.0049	0.0052	0.0136	0.0046	0.0039
Average	0.00058	0.012	0.00628	0.0046	0.09534	0.01214	0.00582
WHO	0.041838	0.316	0.3	0.4	0.998	0.145712	0.978

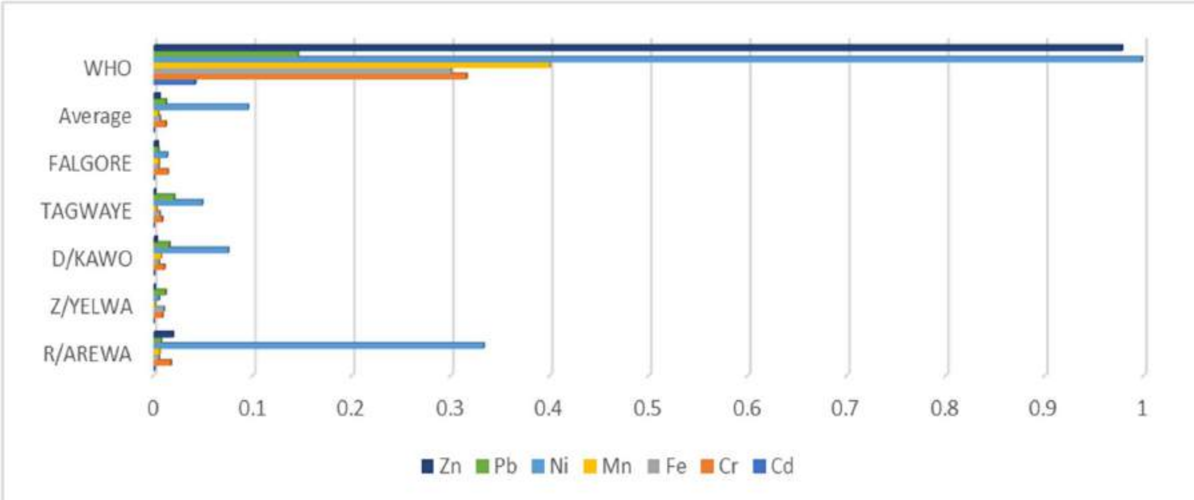

Fig. 15 Comparison of Heavy metal average and Recommended WHO values.

Table V. Chronic daily intake, hazard Quotient, Hazard Index and Life cancer Risk in Adults.

	Element	CDI_{ing}	$CDI_{(derm)}$	$HQ_{(ing)}$	$HQ_{(derm)}$	HI	LCR
	Cd	0.012621	2.88E-05	25.2417	1.151021	0.01265	0.077163
	Cr	0.261121	2.88E-05	522.242	47.62847	0.262312	0.002361
	Fe	0.136653	2.88E-05	273.3066	124.6278	0.139769	0.139769
Adult	Mn	0.100096	2.88E-05	200.1928	91.2879	0.102379	0.102379
	Ni	2.074606	2.88E-05	4149.213	37.84082	2.075552	1.743464
	Pb	0.264167	2.88E-05	528.3348	96.36827	0.266577	0.133288
	Zn	0.126644	2.88E-05	253.2874	69.29942	0.128376	0.128376
	Average	0.42513	2.88E-05	8.502597	6.688625	0.426802	0.3324
	USEPA	B	B	H≤1	H≤1	HI≤1	1.00E-06

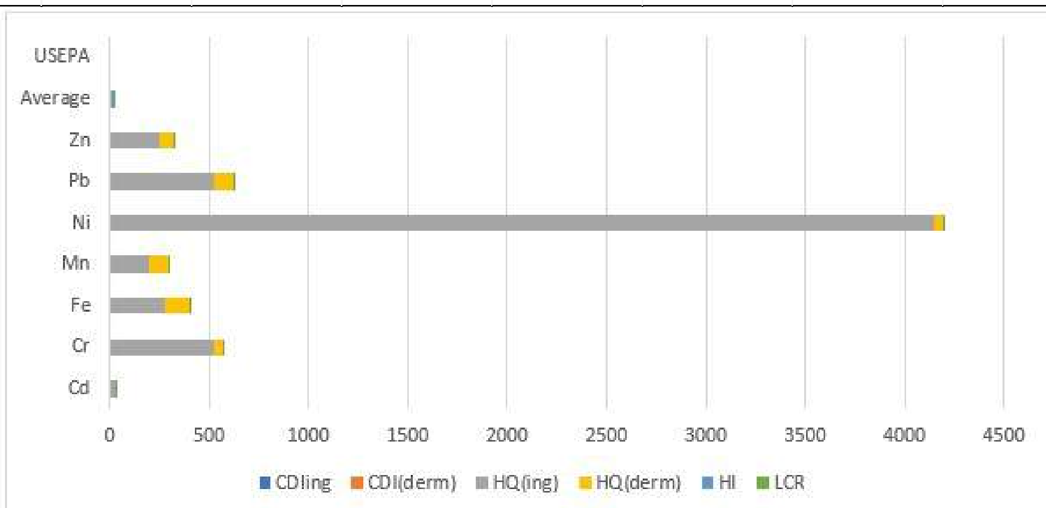

Fig. 16 Comparison of Hazard Quotient, Hazard Index and life cancer risk and Recommended USEPA values.

Table VI. Chronic daily intake, hazard Quotient, Hazard Index and Life cancer Risk on Child.

	Element	CDI_{ing}	CDI_{derm}	HQ_{ing}	HQ_{derm}	HI	LCR
	Cd	0.000185	3.12E-06	0.370898	0.124622	0.000189	0.00115
	Cr	0.003837	1.29E-05	7.67376	0.515677	0.00385	3.46E-05
	Fe	0.002008	3.37E-05	4.015934	1.349354	0.002042	0.002042
Child	Mn	0.001471	2.47E-05	2.941608	0.98838	0.001496	0.001496
	Ni	0.030484	1.02E-05	60.96802	0.409705	0.030494	0.025615
	Pb	0.003882	2.61E-05	7.763287	1.043386	0.003908	0.001954
	Zn	0.001861	1.88E-05	3.721774	0.75031	0.00188	0.00188
	Average	0.006247	1.85E-05	12.49361	0.740205	0.006266	0.004882
	USEPA			H≤0.5	H≤0.5	HI≤0.5	1.00E-04

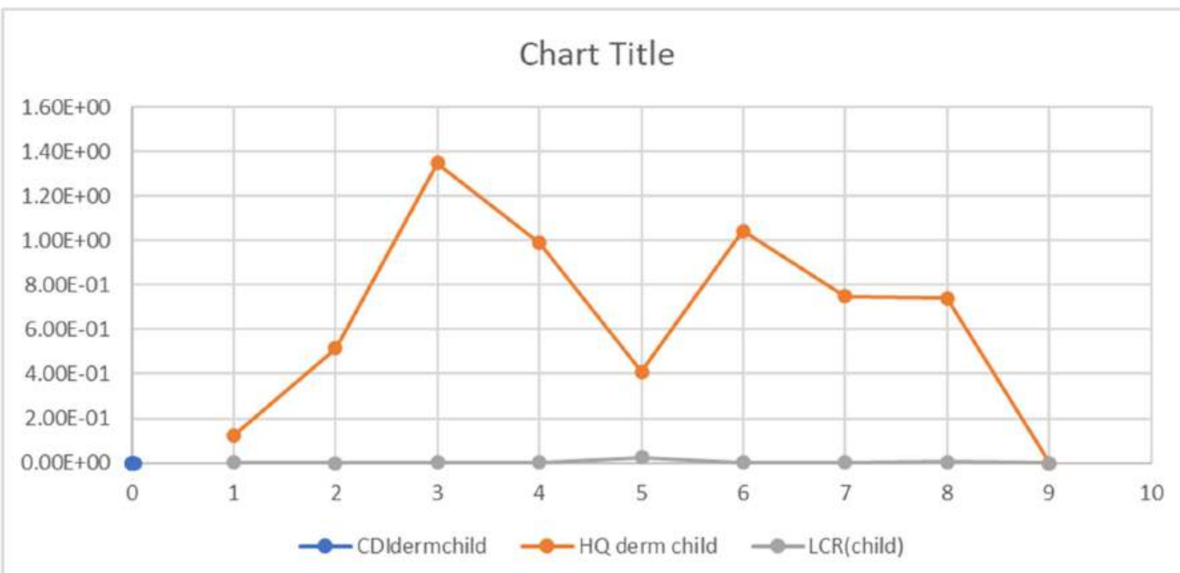

Fig. 17 Trends of chronic daily intake, hazard quotient, and lifetime cancer risk for children.

The average HQ for both ingestion and dermal exposure in children significantly exceeds the recommended value of HQ ≤ 0.5. The LCR value is also higher than the USEPA recommended value of 0.0001, indicating a considerable potential risk. These findings suggest that there is a substantial health risk to children using the water in the area. Fig. 17 illustrates the trends of chronic daily intake, hazard quotient, and lifetime cancer risk for children.

Table VII reveals that the cancer risk from ingestion for adults and children is 0.331069 and 0.004865, respectively. Additionally, the cancer risk from dermal exposure is 0.001331 and 0.000069 for adults and children. The table also presents the lifetime cancer risk for adults and children as 0.3324 and 0.004882, respectively.

Adults face a higher potential risk of cancer compared to children. This difference is attributed to the fact that all mining activities in the areas are carried out by adults. Fig. 18 illustrates the comparison of cancer risk between adults and children, and it also depicts the graphical trends of the cancer risk.

Table VII. Cancer Risk in Adults and Children, Life cancer risk in adults and children.

Element	CR (ing) (Adult)	CR (ing) (Child)	CR(derm) (Adult)	CR(derm) (Child)	LCR (Adult)	LCR (Child)
Cd	0.076987	0.001131	0.000176	1.90E-05	0.077163	0.00115
Cr	0.00235	3.45E-05	1.07E-05	1.16E-07	0.002361	3.46E-05
Fe	0.136653	0.002008	0.003116	3.37E-05	0.139769	0.002042
Mn	0.100096	0.001471	0.002282	2.47E-05	0.102379	0.001496
Ni	1.742669	0.025607	0.000795	8.60E-06	1.743464	0.025615
Pb	0.132084	0.001941	0.001205	1.30E-05	0.133288	0.001954
Zn	0.126644	0.001861	0.001732	1.88E-05	0.128376	0.00188
Average	0.331069	0.004865	0.001331	1.69E-05	0.3324	0.004882

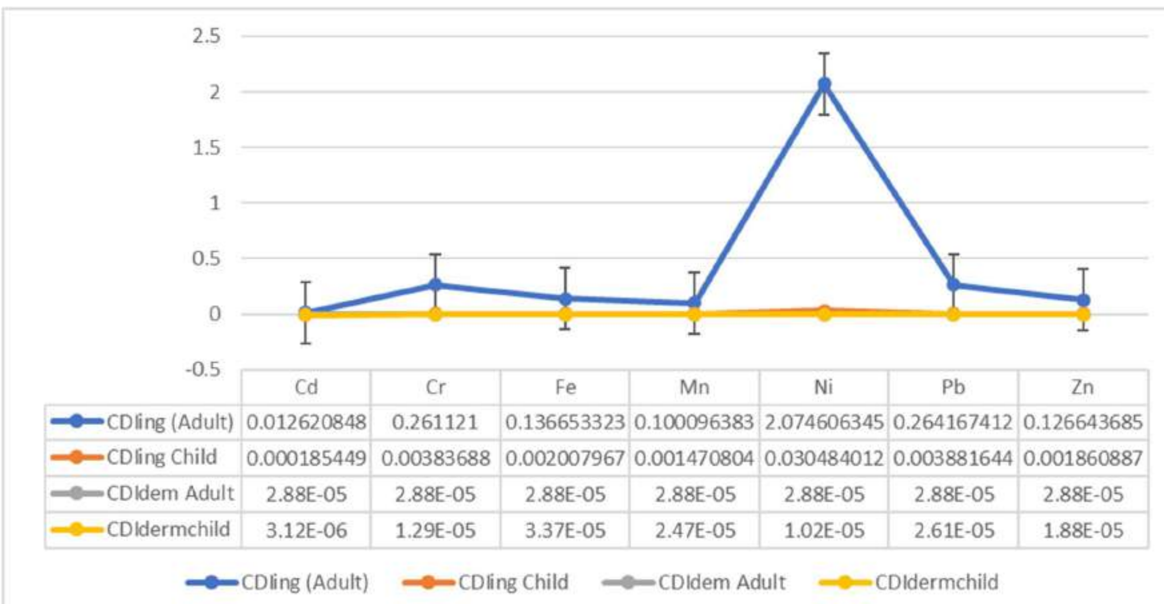

Fig. 18 Comparison of cancer risk between adults and children, and its graphical trends.

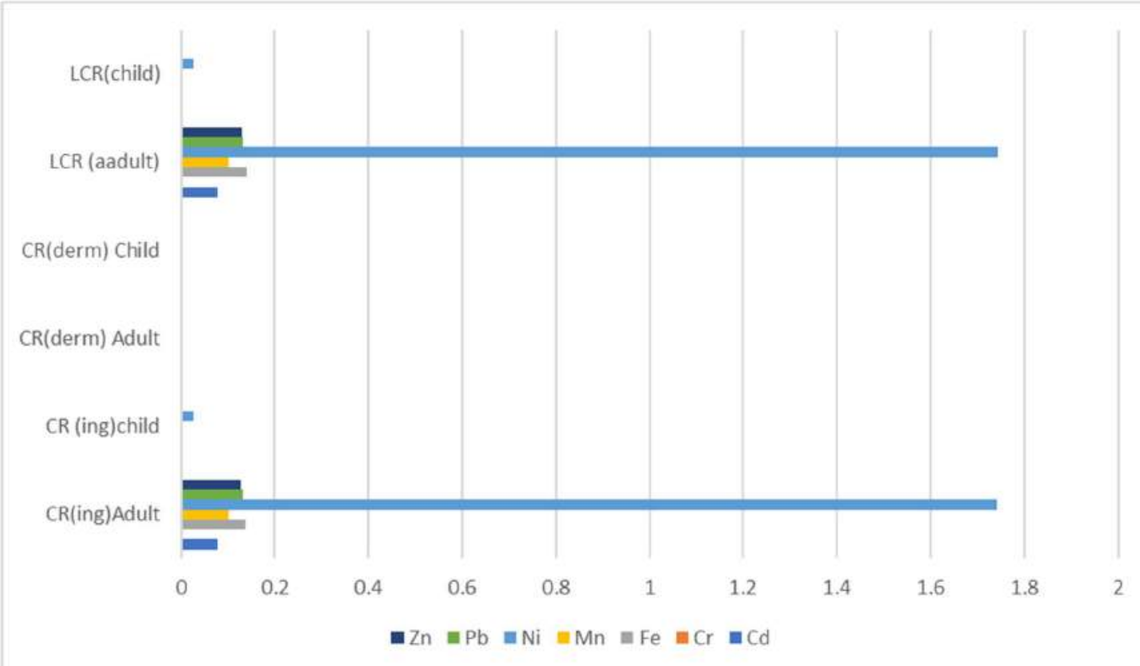

Fig. 19 Comparison of Cancer Risk and Life Cancer Risk in Adults and Children.

IV. Conclusion

This study evaluated the concentrations of gross alpha, gross beta, radon, and heavy metals in groundwater from illegal mining areas in Doguwa Local Government, Kano State, Nigeria, and assessed the associated health risks. The results showed that while gross alpha and radon levels were within World Health Organization (WHO) safety limits, gross beta activity significantly exceeded the recommended limit, posing potential health risks. The average gross beta activity level was 1.7056 Bq/L, above the WHO limit of 1 Bq/L, resulting in annual effective doses from beta radiation exceeding the recommended safety limit of 0.1 mSv/year, particularly for children.

Most heavy metals were within permissible limits, but cumulative exposure to nickel posed a significant health risk, especially to children. The hazard quotient values for children surpassed USEPA recommended thresholds, with a lifetime cancer risk of 0.004882, indicating considerable risk from prolonged exposure.

The findings highlight substantial health risks from both beta radiation and heavy metal contamination in groundwater, particularly the increased risk of cancer and other adverse health effects among children. There's a need to immediately provide clean water, educate the public, install effective water purification systems, offer regular health screenings and treatments, and stop or regulate illegal mining activities to reduce radiological hazards.

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